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Environmental Impact of Concrete and Concrete-Based Construction Waste Leachates

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Abstract. Studies concerning environmental impact of construction materials are very rarely based on experiments performed with real mixed samples and test organisms (ecotoxicological bioassays). In our study, ecotoxicity of two concrete samples from different producers and one concrete-based construction waste in leachate form were assessed. Leachates were treated by various designs, i.e. i) without any treatment, ii) original pH + nutrients addition, iii) pH adjustment to 7.0, and iv) pH adjustment to 7.0 + addition of inorganic nutrients. Ecotoxicological bioassays with freshwater algae *Desmodesmus subspicatus*, freshwater plant *Lemna minor* and freshwater invertebrate *Daphnia magna* were performed. The metal content was determined both in solid samples and leachates. Results showed differences in toxicity level among the concrete sources when original leachates without any treatment were tested. On the contrary, the test design recommended by Czech legislation, i.e. lowering of the pH and addition of nutrients usually significantly decreased the sample toxicity and the potential differences among concrete samples. We suggest that the toxicity of concrete leachates may result not only from the highly alkaline pH but also from the potential persistence of high pH values both within dilution process and time. The higher toxicity was in accordance with higher level of leachate conductivity. Therefore, for the purposes of aquatic ecotoxicity assessment, we recommend either making no pH adjustment or performing the bioassays both with treated and untreated leachates.

1. Introduction

Studies concerning environmental impact of construction materials are usually based on Life Cycle Assessment (LCA) methodology where ecotoxicity is often involved. Nevertheless, the ecotoxic potential is usually simplified and based on raw materials data while experiments with real mixed samples and model organisms, i.e. ecotoxicological bioassays are very rare up to date [1][2].

Building materials and wastes consist of both inorganic and organic compounds. Although the majority of them are not water-soluble, it is necessary to assess their leaching potential and consequent impact on aquatic organisms. One of the possible approaches of ecotoxicity assessment is providing a battery of aquatic bioassays with leachates. Concrete materials generally produce highly alkaline leachates. Upon the Czech legislation, the pH of leachates can be adjusted to 7.8 ± 0.3 prior to the performance of ecotoxicological bioassays [3].

Recently, the partial replacement of natural aggregate using recycled materials originated from concrete waste on one hand and using various chemical components in concrete on the other hand leads to the increased requirement of ecotoxicological assessment of concrete mixtures, where model


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organisms are used. The aim of this study was to determine the suitable method which could be revealing possible ecotoxicological impact of concrete samples from various sources.

2. Materials and Methods

2.1. Samples treatment and leachates preparation style

Three samples were used for the study. Concrete samples (C1 and C2) were obtained from two competitive industrial companies in Czech Republic. Both concrete samples were freshly prepared prior to transport into the laboratory. Concrete waste (W) was collected in DTA Group Praha, Dolní Měcholupy, from the building-waste landfill. W sample represented recycled concrete. Air-dried samples were homogenized and particles up to 10 mm were used for leachate preparation upon [2] in an overhead shaker (7 rpm; 24 h). Consequently, the leachates were centrifuged (4600 rpm, 10 min, 25 °C) and filtered using membrane paper (pores ~ 4 μm). Leachates were left untreated (i) or treated by various designs, i.e. ii) original pH + addition of inorganic nutrients; iii) pH adjustment to 7.0; and iv) pH adjustment to 7.0 + addition of inorganic nutrients**.** Conductivity and pH was determined in the leachates after every treatment or dilution as well as at the end of ecotoxicological bioassays (25 °C, WTW Multiline P4).

2.2. Ecotoxicological bioassays

Ecotoxicological bioassays were carried out first with undiluted leachates for all treatment types (i-iv). In case of treatments i and iv samples were also diluted by control growth media creating a concentration rate of 0 (= control), 6.25, 12.5, 25, 50 and 100 % leachate**.** Addition of inorganic nutrients (treatments ii and iv) and diluting control growth media (treatments i and iv) was always selected upon the test organism. Each sample/control was represented by 3 or 5 replicates, respectively.

2.2.1. Daphnia acute toxicity test. Daphnia mobility inhibition assay was performed using juvenile individuals of *Daphnia magna* Straus aged up to 24 h, originated from ephippia (Microbiotests Inc., Mariakerke (Gent), Belgium). The test design was based on ISO guideline 6341[7]. Aerated ADaM medium (pH ~ 7.8 ± 0.2; $O_2 \geq 7.0$ mg.l^{-1}) according to [6] was used as a control. 5 juveniles were placed into 25-ml-beakers filled with 25 ml of sample / control, covered with transparent film and incubated under stable temperature (24 ± 2 °C) and light cycle (2,000–3,000 lux; 16 h light / 8 h dark). The mobility (viability) of the test organisms was observed after the 48h-exposition.

2.2.2. Freshwater algae toxicity test. Algal growth inhibition test was performed with freshwater green algae *Desmodemus subspicatus* (R. Chodat) E. Hegewald *et* A. Schmidt, strain number Brinkmann 1953/SAG 86.81 (CCALA, IBOT, AS CR, Třeboň, Czech Republic) following the ISO guideline 8692 [7]. BBM media (pH 6.6 ± 0.2) according to [8] was used as control medium. 25-ml-Erlenmeyer flasks filled with 15 ml of sample/control medium were inoculated with 80,000 cells per 1 ml, covered with sterile cellulose and kept under stable temperature (22 ± 2 °C), in constant light (6,000–8,000 lux), and shaking (110 rpm) for 72 h. Algal density was determined by cell counting in a Bürker chamber.

2.2.3. Lemna growth inhibition test. Duckweed assay was proposed by ISO guideline 20079 using *Lemna minor* L., strain Steinberg (FDA, Berlin, Germany), where Steinberg medium (pH 5.5 ± 0.2) served as the control [9]. The test was carried out in 150-ml-beakers, filled with 100 ml of sample/control medium. Each vessel was inoculated with 12 fronds of duckweed of a similar total frond area and covered with transparent film. Test vessels were kept in a stable temperature (24 ± 2 °C) and exposed to a light cycle (5,000–6,000 lux; 16 h light/8 h dark). The total frond area was determined by image analysis using NIS Elements 4.2 (Laboratory Imaging, 2014). Frond area was represented by growth rate values based on repeated measurements three times per exposure, i.e. 0^{th}, 3^{rd} and 7^{th} day. After the 7-day exposure the chlorophyll content was determined by extraction in 98% methanol (48 h; 4 °C, dark) followed by spectrophotometry (Hach, DR/2400, Germany). The calculation of the total chlorophyll content was made according to [11].

2.2.4. Evaluation of ecotoxicological data. Ecotoxicological data for the undiluted samples where expressed as the values of inhibition/stimulation, where tested organisms in samples were compared to control organisms. Results of the tests with concentration rates were demonstrated as values of live daphnia percentage, growth rate (algae, frond area in duckweed), or chlorophyll content per frond area in duckweed.

2.3. Chemical analysis

The metal content in powdered cement was determined by X-ray fluorescence (XRF spectrometer ARL 9400). Metals of low concentration (< 1 %) were determined from acidic extracts using Microwave Plasma-Atomic Emission Spectrometer (Agilent 4200 MP-AES)**.** The acidic extracts were prepared from 5 g of homogenized sample mixed with 20 ml HNO_3, 5 ml 30% H_2O_2 and demineralised H_2O (total mixture volume 100 ml). The mixtures were boiled for 2 hours, filtered and consequently acidified with 5% HNO_3 (total extract volume 200 ml). The quantity of metals in aqueous leachates of the complete concrete samples was measured by Atomic Absorption Spectrometry (AAS, SensAA, GBC).

3. Results and discussion

3.1. Experiments with undiluted leachates

Table 1. Growth inhibition (values 0–100) / stimulation (values < 0) of test organisms under different treatments of undiluted leachates. All values are in % + SD.

	Original pH			Original pH + nutrients		
Organism (endpoint)	C1	C2	W	C1	C2	W
Daphnia	100 ± 0	100 ± 0	100 ± 0	100 ± 0	100 ± 0	100 ± 0
Algae	74 ± 4	76 ± 5	30 ± 1	20 ± 2	85 ± 3	13 ± 0
Duckweed area	100 ± 0	100 ± 0	100 ± 0	100 ± 0	100 ± 0	100 ± 0
Duckweed chlorophyll	100 ± 0	100 ± 0	100 ± 0	100 ± 0	100 ± 0	100 ± 0
	pH adjusted to 7.0			pH adjusted to 7.0 + nutrients		
Organism (endpoint)	C1	C2	W	C1	C2	W
Daphnia	40 ± 0	40 ± 0	100 ± 0	80 ± 0	40 ± 0	100 ± 0
Algae	51 ± 1	63 ± 6	16 ± 4	8 ± 4	7 ± 1	5 ± 2
Duckweed area	45 ± 11	24 ± 15	-8 ± 2	-35 ± 5	-26 ± 5	-47 ± 5
Duckweed chlorophyll	61 ± 8	68 ± 6	38 ± 5	-3 ± 4	42 ± 6	37 ± 4

The untreated leachates were highly alkaline (ranging from 11.6 to 12.3, see table 2) and further addition of nutrients had not significant influence on the pH level (data not shown). The effect of undiluted leachates on test organisms is shown in table 1. The leachates of original pH were found lethal to both daphnia and duckweed (100% inhibition) and further addition of nutrients had no effect. The toxic impact on algae was also high but addition of appropriate salts usually lowered the level of growth inhibition. Recycled concrete (W) was found as the least toxic.

pH adjustment to 7.0 and subsequent nutrient addition significantly lowered the toxic impacts on all organisms with a few exceptions. Although W sample was found as the least toxic for algae and duckweed, it remained lethal to daphnia regardless on the treatment (table 1).

These preliminary tests showed that potential toxicity of the leachates resulted from extreme pH rather than from the lack of nutrients.

3.2. Dose-response experiments

Table 2. Chemical-physical properties of the original and treated leachates (pH adjustment to 7.0 followed by Steinberg salts addition) in duckweed assay. All samples were diluted by control (Steinberg medium). Final pH values were measured after 7-day-test with plants.

Treatment		Original pH			pH adjusted to 7.0 + nutrients		
		Conductivity	pH		Conductivity	pH	
C (%)	Sample	($\mu S.cm^{-1}$)	initial	final	($\mu S.cm^{-1}$)	initial	final
0	Control	977 ± 2	5.5 ± 0.0	5.9 ± 0.1	1020 ± 10	5.5 ± 0.0	6.0 ± 0.3
6.25	C1	856 ± 49	8.6 ± 0.3	6.9 ± 0.0	1109 ± 7	5.9 ± 0.1	6.1 ± 0.0
	C2	880 ± 28	8.4 ± 0.3	8.2 ± 0.1	1145 ± 1	5.8 ± 0.0	6.4 ± 0.1
	W	829 ± 14	7.1 ± 0.3	7.1 ± 0.0	1094 ± 13	5.9 ± 0.0	6.0 ± 0.1
12.5	C1	876 ± 59	10.4 ± 0.1	7.5 ± 0.0	1195 ± 16	5.8 ± 0.0	6.2 ± 0.1
	C2	988 ± 13	10.3 ± 0.3	9.7 ± 0.0	1287 ± 7	5.9 ± 0.0	6.3 ± 0.0
	W	816 ± 20	7.7 ± 0.4	7.4 ± 0.0	1105 ± 4	6.0 ± 0.0	6.2 ± 0.1
25	C1	1017 ± 68	11.4 ± 0.1	8.4 ± 0.1	1373 ± 17	6.0 ± 0.1	6.4 ± 0.0
	C2	1373 ± 18	11.4 ± 0.2	11.2 ± 0.0	1567 ± 4	6.1 ± 0.0	6.4 ± 0.1
	W	751 ± 8	8.3 ± 0.3	8.1 ± 0.1	1190 ± 2	6.1 ± 0.1	6.5 ± 0.1
50	C1	1487 ± 92	11.9 ± 0.0	10.3 ± 0.0	1735 ± 44	6.2 ± 0.1	6.6 ± 0.1
	C2	2330 ± 28	11.9 ± 0.1	11.7 ± 0.0	2125 ± 7	6.3 ± 0.0	6.6 ± 0.3
	W	780 ± 6	11.0 ± 0.3	10.2 ± 0.1	1376 ± 34	6.3 ± 0.1	6.9 ± 0.1
100	C1	2575 ± 41	12.2 ± 0.1	11.6 ± 0.0	2443 ± 95	6.5 ± 0.2	6.9 ± 0.1
	C2	4300 ± 71	12.3 ± 0.1	12.1 ± 0.2	3150 ± 14	6.5 ± 0.0	6.4 ± 0.4
	W	1075 ± 40	11.6 ± 0.1	11.5 ± 0.0	1675 ± 6	6.6 ± 0.0	7.6 ± 0.1

As table 2 shows, the initial pH values of the untreated diluted leachates remained relatively high for both concrete samples (C1, C2) whereas the decrease in the waste sample (W) occurred faster. This phenomenon could be related with the conductivity, where higher conductivity level lead to higher pH of the diluted samples. The initial pH of the untreated leachates remained quite stable to the end of exposition in C2 and W samples, whereas significant decrease in pH after 7 days was found in original C1 leachate.

In algal test, the initial as well as the final pH values of untreated samples were also relatively high. The increased pH values might result from the photosynthetic activity and CO_2 depletion in the test media [12]. Therefore, algae represent a good test organism for concrete leachates ecotoxicity assessment due to their relative resistance to alkaline environment (figure 2). On the contrary, daphnia are highly sensitive both to extreme pH and high conductivity values which resulted in the highest ecotoxicological response. The original leachates were highly toxic (W) or lethal (C2) for daphnia and the treated leachates also showed differences among the samples (figure 1). Duckweed was seen also relatively sensitive to the studied samples. Treated leachates lead to frond growth stimulation on one hand and a slight chlorosis at the same time (figure 3, 4). The highest ecotoxicological impact found in untreated C2 leachate was probably connected with the highest values of both conductivity and pH (table 2).

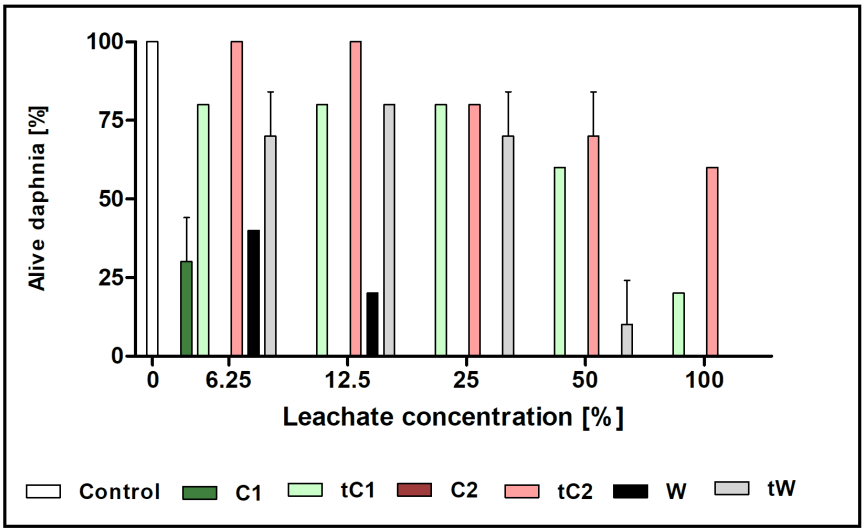


Figure 1. Effect of concrete leachates on daphnia viability: dose-response relationships. C1, C2 and W... untreated leachates (orginal pH); tC1, tC2 and tW... treated leachates (pH adjusted to 7.0 + ADaM nutrients addition). All samples diluted by control (ADaM medium). Values for C2 leachate were zero in all the concentration range.

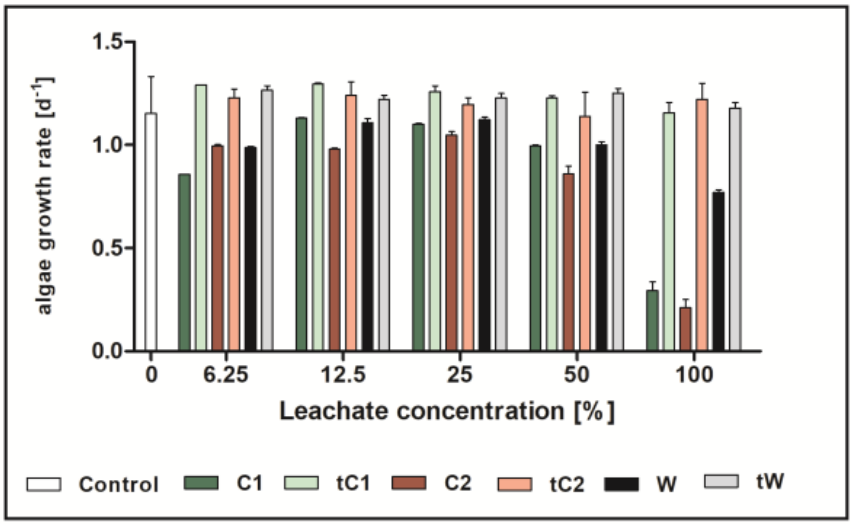


Figure 2. Effect of concrete leachates on algal growth rate: dose-response relationships. C1, C2 and W... untreated leachates (orginal pH); tC1, tC2 and tW... treated leachates (pH adjusted to 7.0 + BBM nutrients addition). All samples diluted by control (BBM medium).

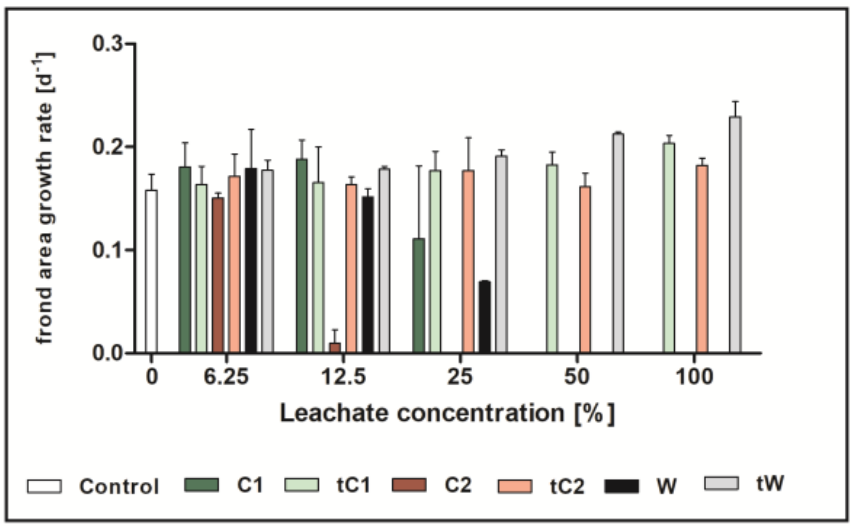


Figure 3. Effect of concrete leachates on duckweed growth rate: dose-response relationships. C1, C2 and W... untreated leachates (orginal pH); tC1, tC2 and tW... treated leachates (pH adjusted to 7.0 + Steinberg nutrients addition). All samples diluted by control (Steinberg medium).

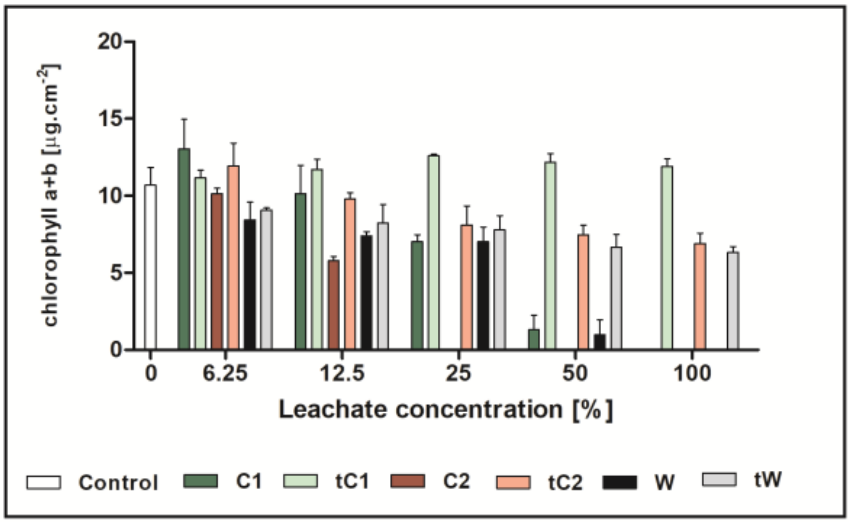


Figure 4. Effect of concrete leachates on duckweed chlorophyll content: dose-response relationships. C1, C2 and W... untreated leachates (orginal pH); tC1, tC2 and tW... treated leachates (pH adjusted to 7.0 + Steinberg nutrients addition). All samples diluted by control (Steinberg medium).

The public notice No. 294/2005 in Czech legislation allows adjusting the pH of the leachates prepared of waste samples when original values are extreme for the test organism; the recommended value is 7.8 ± 0.3 [3]. This is in accordance with the international guidelines for ecotoxicity testing, where pH value of the test sample should be the same as the pH level of the control media [5][7][9]. In our experiment we have chosen a compromise pH value (7.0) which lies within the range of control media pH for the selected test organisms (5.5–7.8). However, such treatment inevitably leads to change of chemical composition of the liquid samples, as well as to change of the toxicity.

Our results showed that testing of the concentration rate of untreated samples revealed both the dose-response relations and different ecotoxicological effect among the samples tested. We suggest that the potential toxicity of concrete leachates consist not only in highly alkaline pH but also in the persistence of the extreme pH values both within dilution process and time. Such extreme pH may lead to negative impact on ground-, soil- or freshwater and so far should not be neutralized, as it is said e.g. in duckweed assay guideline [9]. Duckweed is tolerant to pH in range of 5.0–9.0, which makes it a suitable test organism for alkaline samples. We also recommend performing the duckweed assay where toxic effects can be observed on morphological and biochemical level at the same time and so far it gives more complex response of the test organism. [9]

3.3. Metal content of the solid samples and leachates

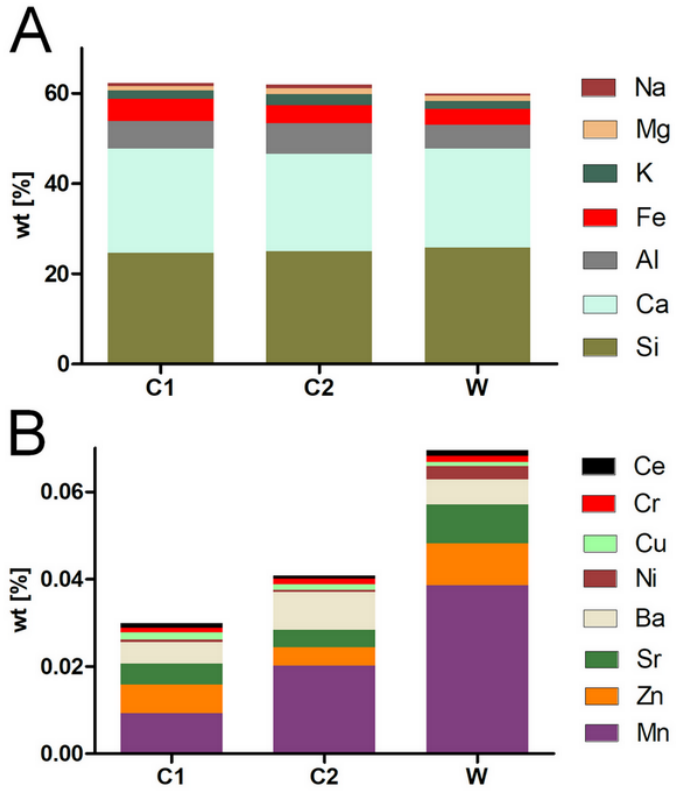


Figure 5. Metals content in the solid samples. Major elements were measured by X-ray fluorescence (A). Minority metals were estimated by AES in acidic extracts (B). Cd and Pb were below detection limit (< 0.4 mg.kg^{-1}).

Modern concrete often contain various admixtures as fly ash that improve the properties of both fresh and hardened concrete. Despite its positive properties, fly ash contains also trace amounts of toxic metals [10]. In our study analysis of the major metals in the solid samples did not show any significant difference (figure 5-A). The toxic metals (Cd, Pb) were below detection limit; Cr ranged in 10.6–14.4 mg.kg^{-1} (figure 5-B). No toxic metals were leached into water eluates. The only element detected in the leachates was calcium (Ca); the highest content (8.3 ± 0.1mg.l^{-1} ~ pH 11.6) was found in W sample. Therefore, the ecotoxic potential of the samples is likely to result from some other leaching components, as ions or water-soluble organic compounds.

4. Conclusion

Our experiments showed different levels of aquatic ecotoxicity among the used model concrete samples. Original leachates without any adjustment were found highly alkaline which had lethal impact on most of the test organisms. However, the differences among the samples were pronounced in dose-response tests with the untreated leachates. In accordance with the results, we conclude that

not only the high value of pH itself but also the level of alkaline pH persistence in both dilution process and time makes the important part of concrete ecotoxicity in leaching systems. Lowering of the pH leads to strong changes of physical-chemical properties of the leachate, reduction of the potential toxicity and decrease in the variability among tested samples. Therefore, when ecotoxicity assessment of virgin or recycled concrete is based on aquatic leachates, we recommend either making no pH adjustment or performing the bioassays both with treated and untreated leachates at the same time. Further research will be conducted with more concrete samples containing both natural and recycled components.

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